Structure and Electron Localization of Reduced Ceria Surfaces
RENAT SABIRIANOV, Univ of Nebraska - Omaha, KHALDOUN TARAWNEH, Princess Sumaya University for Technology, NABIL AL-AQTASH, NAN SHAO, WAI-NING MAI, Univ of Nebraska - Omaha, CHIN LI CHEUNG, Univ of Nebraska Lincoln — Ceria (CeO$_2$) is an extensively used industrial catalyst. However, the mechanism for its catalytic activity, especially in aqueous media, is not yet well-understood. While high density of oxygen vacancy defects (OVDs) are often cited as the major factor in enhancing the activity of nanostructured ceria, the synergistic influences between the surface defects, subsurface defects and internal defects has not been explored systematically. Using density-functional theory (DFT) with DFT+U approach, we study the local structure of surface and subsurface oxygen vacancies on the (100), (111) and (110) surfaces of ceria. The removal of a neutral surface oxygen atom leaves back excess electrons that are shown to localize on cerium ions neighboring the defect. The preferential defect formation and the different chemical reactivity of the (100), (111) and (110) surfaces are discussed in terms of defect formation energies. DFT+U calculations predict the preferential subsurface formation of OVDs for (111) surface by energy difference of 0.10 eV, in agreement with previous GGA+U results, while in case of (110) surface calculations predict the surface vacancy to be more stable by energy difference of 1.03 eV. The calculated atomistic and electronic structures of ceria the reduced surfaces are shown to agree with spectroscopic and STM measurements.

Renat Sabirianov
Univ of Nebraska - Omaha

Date submitted: 14 Nov 2014
Electronic form version 1.4